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## Report Title

Final Report: Coherent Preparation of Molecular Hydrogen in (v,J,M) Eigenstates for Reaction Dynamics Studies

### ABSTRACT

We have proposed and demonstrated a robust coherent optical technique, the “Stark induced adiabatic Raman passage” or SARP for preparing single vibrational energy eigenstate of a molecule. Using SARP we have, for the first time, demonstrated population inversion between a ground vibrational ( $v=0, J$ ) and an excited ( $v > 0, J'$ ) vibrational level of H<sub>2</sub> within the ground X  $1^2\Sigma^+$  electronic state. In addition, we have prepared a coherent superposition of degenerate M-states belonging to a single rovibrational ( $v=1, J=2$ ) energy eigenstate within the ground X  $1^2\Sigma^+$  electronic state of H<sub>2</sub>. The phase entangled superposition states opened new avenues to coherently control the collision dynamics, which may also allow us to measure the geometrical phase effect in a collision process. SARP provides a unique way to prepare high lying vibrational energy eigenstate using commercially available nanosecond pump and Stokes pulses.

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**Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:**

#### (a) Papers published in peer-reviewed journals (N/A for none)

Received

Paper

08/28/2014 1.00 Wenrui Dong, Nandini Mukherjee, Richard N. Zare. Optical preparation of H<sub>2</sub> rovibrational levels with almost complete population transfer, The Journal of Chemical Physics, (08 2013): 0. doi: 10.1063/1.4818526

08/28/2014 2.00 Richard N. Zare, Wenrui Dong, Nandini Mukherjee. Coherent superposition of M-states in a single rovibrational level of H<sub>2</sub> by Stark-induced adiabatic Raman passage, The Journal of Chemical Physics, (02 2014): 0. doi: 10.1063/1.4865131

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Number of Papers published in non peer-reviewed journals:

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**Non Peer-Reviewed Conference Proceeding publications (other than abstracts):**

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Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Richard N. Zare	0.01	Yes
Nandini Mukherjee	1.00	No
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# Coherent Preparation of Molecular Hydrogen in (v, J, M) Eigenstates for Reaction Dynamics Studies

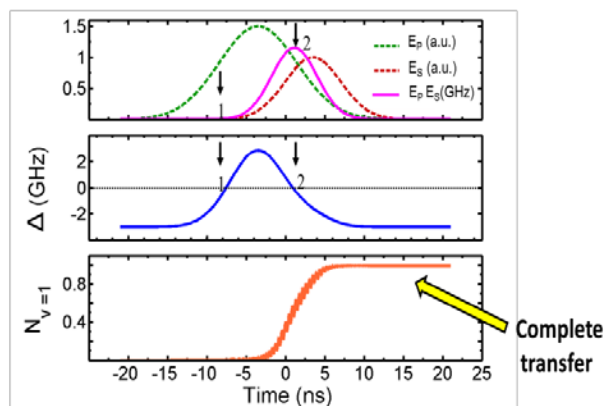
Nandini Mukherjee, William E. Perreault, Christian Chamberlayne, and Richard N. Zare

To understand a chemical reaction such as  $\text{H}_2 + \text{D} \rightarrow \text{HD} + \text{H}$  at the most fundamental level it is essential to prepare the target molecule ( $\text{H}_2$ ) in an addressable vibrational rotational quantum state. The challenge here is to transfer a substantially large number of the ground state  $\text{H}_2$  molecules into a desired target quantum state so we can study bimolecular collision dynamics with appreciable signal to noise. Under the auspices of ARO support (Grant W911NF-13-1-0126) we have successfully carried out this project preparing a target molecule ( $\text{H}_2$ ) in a single or in a superposition of M states within a single rovibrational (v, J) level of the  $\text{H}_2$  molecule (or its isotopic forms, HD and  $\text{D}_2$ ) within its ground  $X^1\Sigma_g^+$  electronic state.

To prepare a vibrationally excited (v, J, M) single quantum state, we introduced a new coherent optical technique called the *Stark induced adiabatic Raman passage* (SARP) [*J. Chem. Phys.* **135**, 024201]. Figure 1 describes the basic mechanism by which SARP achieves the complete population transfer to a target quantum state.

## Stark-Induced Adiabatic Raman Passage (SARP)

A strong pump followed by weaker Stokes pulse (or, vice versa) transfers **all** population to desired quantum state



**Figure 1.** Simulation of SARP showing the complete population transfer from  $\text{H}_2$  ( $v=0, J=0$ ) to  $\text{H}_2$  ( $v'=1, J'=2$ ) using partially overlapping nanosecond pump,  $E_P$  and Stokes,  $E_S$  pulses.

As shown in Fig. 1, to achieve the complete population transfer to a desired target state SARP utilizes a sequence of partially overlapping off-resonant nanosecond pump ( $E_P$ ) and Stokes ( $E_S$ ) pulses of unequal intensities (upper panel of Fig. 1). The pulse with the higher intensity (pump or Stokes) generates the necessary sweep of the Raman resonance frequency by inducing the second-order (dynamic) Stark shifts of the upper and lower rovibrational levels. During pulsed excitation the Raman transition frequency is swept through resonance twice as shown by the arrows in the middle panel of Fig. 1. The delay between the two pulses is adjusted so that only one of these crossings (indicated by the second arrow) is avoided in the presence of a strong Raman coupling (overlap of the pump,  $E_P$  and Stokes,  $E_S$  pulses). As the Raman resonance is crossed with a sufficiently strong Raman coupling, a unidirectional flow of population from the initial to the final target state takes place during the overlap of the pump and Stokes pulses as shown in the lower panel of Fig. 1.

To excite the vibrational rotational levels of  $H_2$  SARP requires a single-mode nanosecond pump laser pulse (1064, 532, 355 nm) with an energy  $\sim 100$ -200 mJ and a single-mode nanosecond Stokes laser pulse (600-700 nm) with an energy  $\sim 10$ -20 mJ. The previous DURIP equipment grants were utilized to acquire the various laser sources such as an injection-locked  $Nd^{+3}$ :YAG laser which provides the pump laser pulses, a single-mode frequency stabilized Matisse dye laser and a seeded pulsed dye amplifier which provides the Stokes laser pulses.

### **Experimental demonstration of SARP transferring the complete population of the ground ( $v=0$ , $J=0$ ) level of $H_2$ to the vibrationally excited ( $v=1$ , $J=0$ ) level within its ground $X^1\Sigma_g^+$ electronic state**

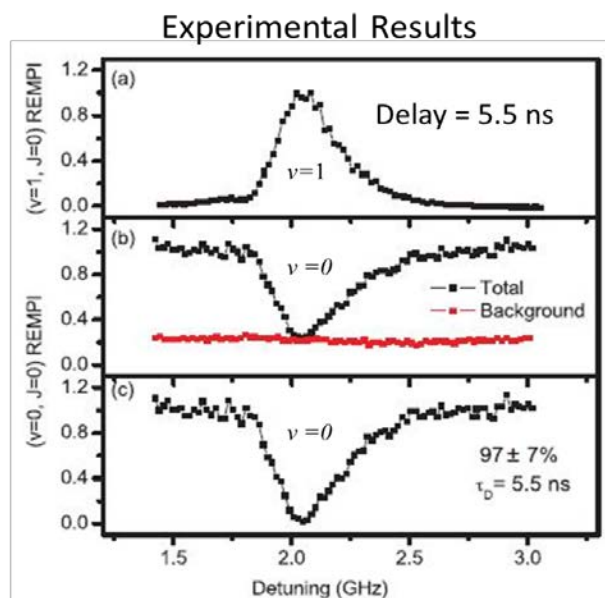
By combining a single-mode pump laser pulse (532 nm, 6 ns) with a single-mode Stokes laser pulse (683 nm, 4.6 ns) with a relative delay of 4-6 ns, SARP was able to transfer the entire population of  $H_2$  ( $v=0$ ,  $J=0$ ) to  $H_2$  ( $v=1$ ,  $J=0$ ). The delayed sequence of the pump and Stokes laser pulses transversely intersected a supersonically expanded molecular beam of  $H_2$  within a high-vacuum reaction chamber. Following SARP excitation,  $H_2$

molecules were probed state selectively using (2+1)

$E, F^1\Sigma_g^+(v'=0, J'=J) - X^1\Sigma_g^+(v=0, 1, J)$  resonance enhanced multiphoton ionization (REMPI).

Figure 2 shows the experimental results achieving the complete population transfer from  $H_2(v=0, J=0) \rightarrow H_2(v=1, J=0)$ . The top panel in Fig 2 shows the (2+1) REMPI signal from the vibrationally excited ( $v=1$ ) level. The population transfer is calibrated from the depletion of the ( $v=0$ ) REMPI signal ((b) and (c) of Fig. 2). The data in Fig. 2 clearly demonstrate that the near complete population transfer from  $H_2(v=0) \rightarrow H_2(v=1)$  has occurred when the background is properly subtracted (lowest panel). This work is published in two papers [*J. Chem. Phys.* **138**, 051101 (2013), and *J. Chem. Phys.* **139**, 074204 (2013)].

SARP pumping of  $H_2(v=0, J=0, M=0) \rightarrow H_2(v=1, J=0, M=0)$



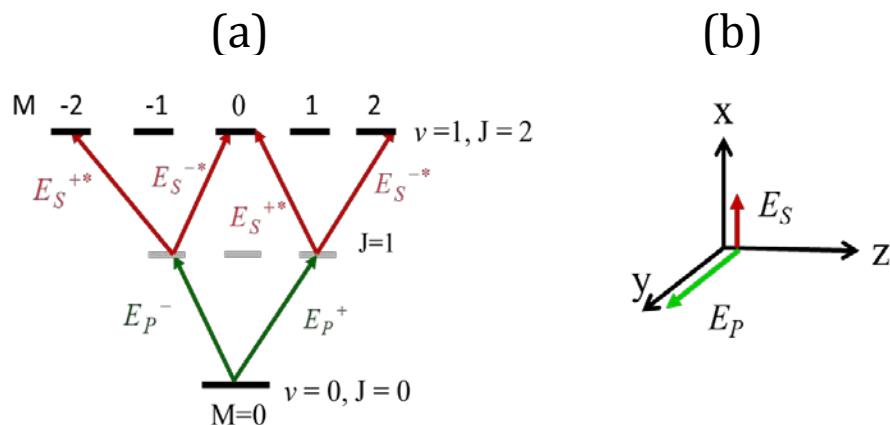
**Figure 2.** Demonstration of SARP achieving the complete population transfer from  $H_2(v=0, J=0) \rightarrow H_2(v=1, J=0)$

## Preparation of a bi-axial superposition state within a single rovibrational $H_2(v=1, J=2)$ eigenstate

By combining different polarizations of the pump and Stokes laser pulses SARP prepared a bi-axial coherent superposition of quantum states belonging to a single ( $v=1, J=2$ )



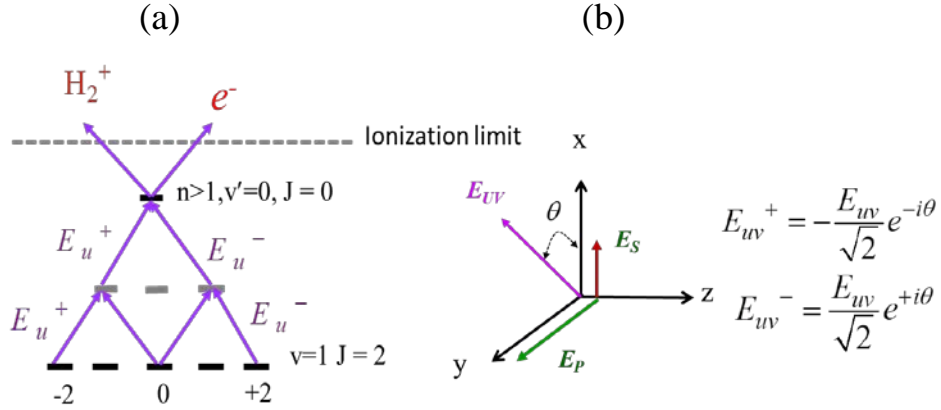
rovibrational energy eigenstate within the ground  $X^1\Sigma_g^+$  electronic state of  $H_2$ . SARP was able to transfer 60% population of  $H_2$  ( $v=0, J=0, M=0$ ) state to the coherent superposition state  $|\psi(t)\rangle = 1/\sqrt{(2)}[|v=1, J=2, M=-2\rangle - |v=1, J=2, M=+2\rangle]$ ; this work has been published [*J. Chem. Phys.* **140**, 074201 (2014)]. The superposition state was prepared with linearly but cross-polarized single-mode pump (532 nm,  $\sim 6$  ns, 200 mJ) and Stokes (699 nm,  $\sim 4$  ns, 20mJ) laser pulses co-propagating along the quantization z-axis as shown in Fig. 3b. Figure 3a describes the excitation scheme using cross-polarized pump and Stokes laser pulses.



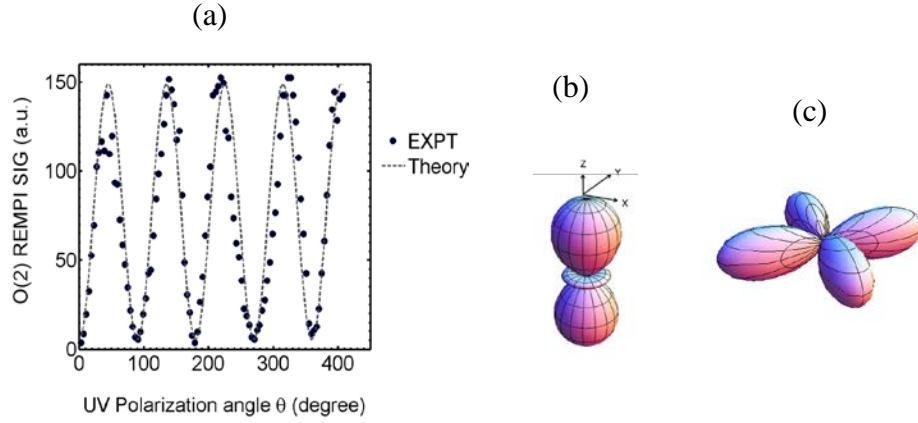
**Figure 3.** (a) SARP excitation scheme to prepare  $M$ -sublevel superposition using left and right circularly polarized pump and Stokes laser pulses. The left  $E^+$  and right  $E^-$  circularly polarized components of the optical fields are derived from the linearly polarized transverse pump and Stokes lasers. (b) Polarization of the pump and Stokes fields with respect to the z-axis oriented along the laser propagation.

We have measured the degree of phase coherence by using the O (2) line of the  $H_2$  E,F-X (0,1) band via 2+1 resonance enhanced multiphoton ionization (REMPI). The 2+1 REMPI excitation scheme using polarized UV probe laser pulses is shown in Fig.4a. The interference fringes of the REMPI generated ion signal (see Fig.5.) are recorded using a time-of-flight mass spectrometer as the direction of the UV laser polarization is rotated using a half-wave plate. The fringe contrast of the REMPI generated ion signal in Fig. 5a directly measures the phase coherence of the superposition state. Figure 5a and b show the distributions of the angular momentum and the rotor axes for the prepared vibrationally excited superposition state. By measuring the depletion of the REMPI

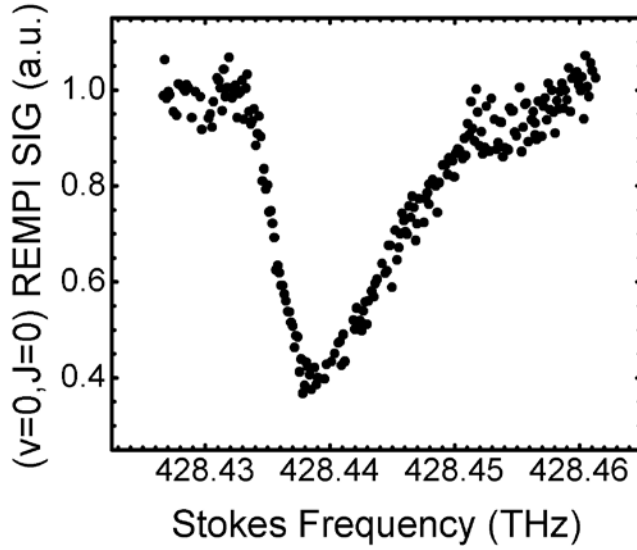
signal from  $H_2$  ( $v=0, J=0$ ) level as the Stokes frequency is tuned across the ( $v=0, J=0$ )  $\rightarrow$  ( $v=1, J=2$ ) Raman resonance, we confirmed that 60% population has been transferred from  $H_2$  ( $v=0, J=0$ ) to the superposition state in  $H_2$  ( $v=1, J=2$ ) (see Fig. 6).



**Figure 4.** (a) 2+1 O(2) REMPI excitation scheme to detect  $M$ -sublevel coherence using polarized UV laser pulses. (b) Polarization direction of UV laser optical field relative to the direction of the pump and Stokes laser field. All laser beams propagate parallel to the quantization  $z$ -axis.



**Figure 5.** (a)  $E, F^1\Sigma_g^+(v'=0, J'=0) \leftarrow X^1\Sigma_g^+(v=1, J=2)$  O(2) REMPI signal from  $H_2$  ( $v=1, J=2$ ) excited state prepared by SARP with cross polarized pump and Stokes laser pulses. The REMPI signal is plotted against the polarization direction (angle  $\theta$ ) of the UV laser relative to the direction of the Stokes polarization (x). (b) 3-D polar plot of the angular momentum polarization with alignment parameters  $A_0^{(2)} = 1$  and  $A_{\pm}^{(2)} = 0$ , calculated using the fitted values of the M-state amplitudes. (c) Bi-axial distribution of rotor axes

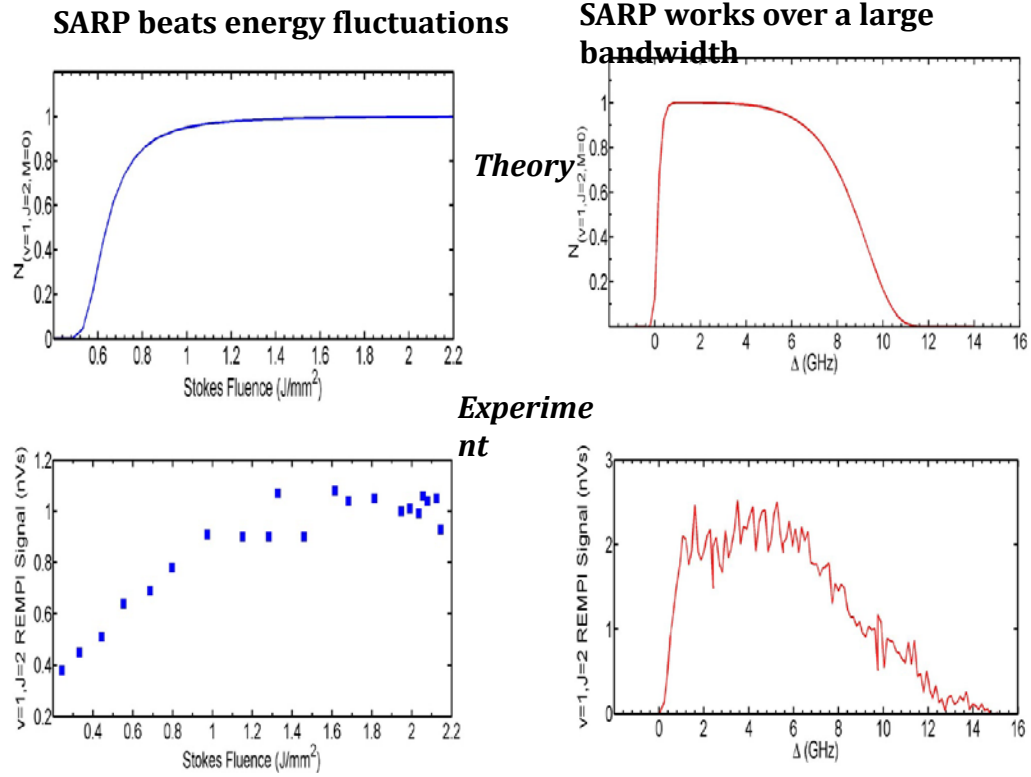


**Figure 6.** Depletion of the Q(0) branch of  $E, F^1\Sigma_g^+(v'=0, J'=0) \leftarrow X^1\Sigma_g^+(v=0, J=0)$

REMPI signal as a function of Stokes laser frequency in THz. The depletion of REMPI signal calibrates population transfer from the ground  $H_2(v=0, J=0) \rightarrow H_2(v=1, J=2)$  level

### **Demonstration of SARP as a robust technique for preparing a desired rovibrational M-quantum state**

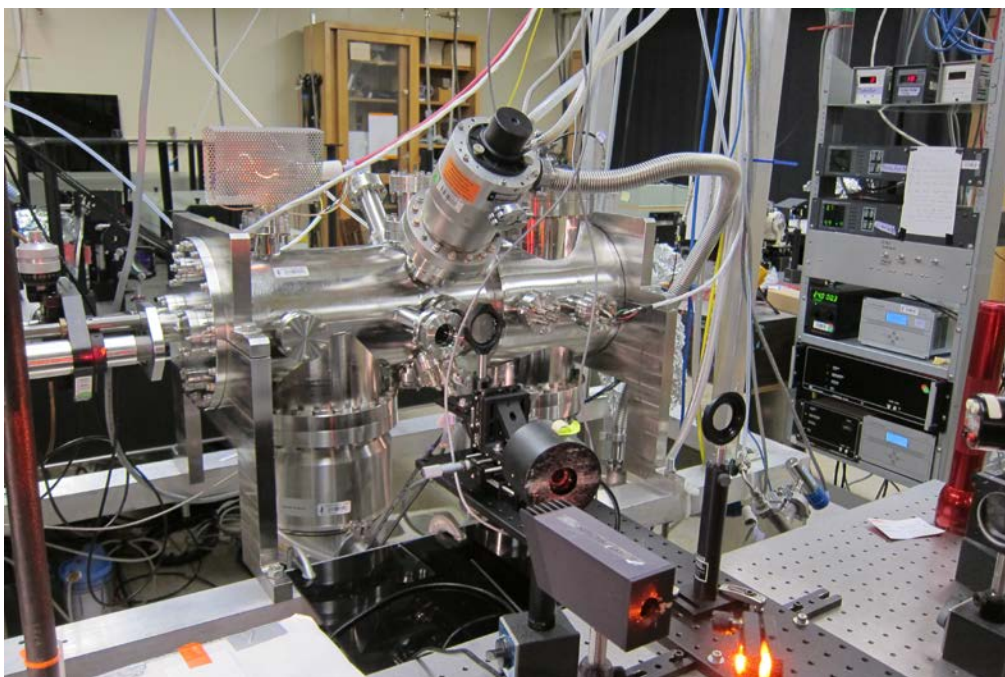
We have also successfully prepared the various vibrationally excited ( $v=1$ ) quantum states of  $H_2$  including ( $v=1, J=1, M=0, \pm 1$ ),  $H_2(v=1, J=3, M=0, \pm 1)$ ,  $H_2(v=1, J=2, M=0, +2 \text{ or } -2)$ . As shown in Figure 7, the SARP induced population transfer to each of these vibrationally excited quantum states demonstrates remarkable stability against energy and frequency fluctuations of the laser pulses, proving SARP to be a robust technique for preparing a desired rovibrational M-quantum state using commercially available laser sources.



**Figure 7.** Theory and experiment demonstrating large bandwidth and stability against laser frequency and power fluctuations for the preparation of the rovibrationally excited  $\text{H}_2$  ( $v=1$ ,  $J=0,1,2,3$ ) states using SARP.

### **Fabrication and calibration of a Time-of-Flight Spectrometer**

To study collisions using target molecules in SARP prepared quantum states we have fabricated a high-vacuum reaction chamber, which combines a collimated supersonic molecular beam with a high-resolution time-of-flight spectrometer. The chamber is differentially pumped and a narrow supersonic molecular beam is extracted and led into the reaction part of the vacuum chamber by using a 0.5 mm skimmer. To obtain a high resolution velocity map imaging of the scattered angular distribution, the time-of-flight spectrometer is equipped with a 25 cm flight tube coupled with a pair of 12 cm diameter multichannel plates (MCP) and a two-dimensional X-Y delay-line anode detector from *RoentDek*. The molecular beam time-of-flight spectrometer is shown in Figure 8.



**Figure 8**

### **Measurement of anisotropy parameter $\beta$ using the time-of-flight-spectrometer**

We have carefully calibrated the time of flight spectrometer by measuring the angular distribution of the ions generated by resonantly enhanced multi-photon ionization (REMPI) with pulsed UV lasers. The UV laser pulse is generated in two steps, via the third harmonic generation of a tunable pulsed dye laser in two  $\alpha$ -BBO crystals. The

pulsed dye laser is in turn pumped by the second harmonic of a Q-switched Nd<sup>+3</sup>:YAG laser from *Continuum*.

In the process of calibrating the time-of-flight-spectrometer we have also measured the anisotropy parameter  $\beta$  for the angular distribution of the photoelectron and photoion in (2+1) resonance enhanced multiphoton ionization process of  $\text{H}_2 \text{ X } ^1\Sigma_g^+$  ( $v=0, J=0$ ) molecules through the intermediate  $\text{H}_2 \text{ E,F } ^1\Sigma_g^+$  ( $v'=0, J'=0$ ) level ( $\lambda = 201.684 \text{ nm}$ ). The time-of-flight spectra were recorded as the direction of polarization of the ionizing laser was varied with respect to the flight axis and were fitted to an angular distribution in an appropriately rotated coordinate system with the z-axis oriented along the time-of-flight axis. The anisotropy parameter  $\beta$  was found to be  $1.72 \pm 0.13$  by fitting the time-of-flight spectra and agreed with previous measurements. Using secondary ionization with a delayed laser pulse of different wavelength we have also determined the vibrational energy distribution of the ions, showing that  $98 \pm 4\%$  of the ions are generated in their ground vibrational state, in agreement with the calculated Franck-Condon factors between the  $\text{H}_2 \text{ E,F } ^1\Sigma_g^+$  ( $v'=0$ ) and  $\text{H}_2^+ \text{ X } ^1\Sigma_g^+$  ( $v''$ ) levels This work has been published [*JCP* 144, 214201 (2016)]

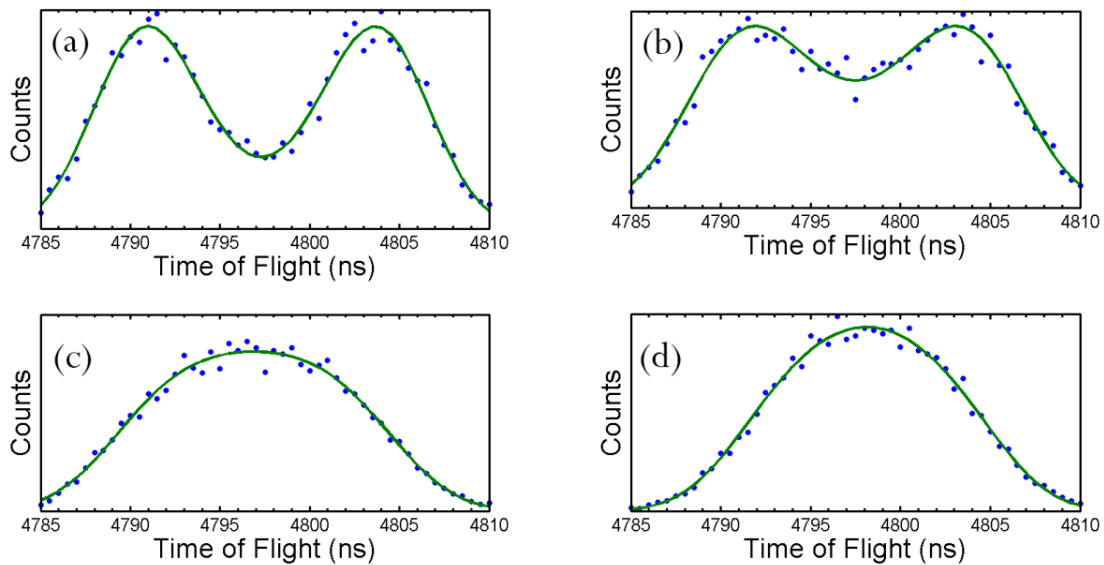


FIG. 9. Time-of-flight spectra of the  $\text{H}_2^+$  ion for (a)  $\theta = 0^\circ$ ; (b)  $\theta = 30^\circ$ ; (c)  $\theta = 60^\circ$ ; and (d)  $\theta = 90^\circ$ . The green curves are fits to the blue data points, based on the fitting for  $\theta = 0^\circ$  using Eq. (6) with  $\alpha = 8.3$  and  $\beta = 1.72$ .

Figure 9 shows the fitted time-of-flight spectra as a function of probe laser polarization. For the polarization direction parallel to the detection axis ( $\Theta = 0^\circ$ ), the recoil effect is most pronounced, and the distribution separates into two peaks corresponding to two possible directions of the ejected electron travelling parallel or anti-parallel to the detection axis. This behavior is clearly evidenced in the top panel Fig. 9a. Starting from the known angular distribution  $I(\theta) = \frac{\sigma}{4\pi}[1 + \beta P_2(\cos\theta)]$  of the photoions with respect to the polarization direction of the ionizing laser, we have derived the following mathematical expression for the time-of-flight-distribution of the photo-ions for a given polarization direction  $\Theta$  of the UV laser with respect to the detection axis:

$$f(\tau) \propto \int_0^\pi (\tau - \alpha \cos \theta')^2 \exp\{ -[(\tau - \tau_o - \alpha \cos \theta') / \Delta\tau]^2 \} \times [1 + \beta P_2(\cos \Theta) P_2(\cos \theta')] \sin \theta' d\theta' \quad (1)$$

Here  $\tau_o$  corresponds to the time-of-flight (TOF) of the peak velocity and  $\Delta\tau$  is the width of the distribution in the absence of photoelectron recoil. The parameter  $\alpha$  is determined by the product of the maximum recoil speed of the photoelectron and the resolution of the TOF mass spectrometer. Eq. (1) was fitted to the TOF spectra of Fig. 9 using the “NonlinearModelFit” program of Mathematica, which produced a value of  $\beta = 1.72 \pm 0.13$ . The  $R^2$  value of the fit was better than 0.99. This large value of the anisotropy parameter corresponds to a highly polarized ejection of the electron in the direction of the optical field.

Figures 9b, 9c, and 9d show the TOF spectra of  $H_2^+$  for the polarization directions  $\Theta = 30^\circ$ ,  $60^\circ$ , and  $90^\circ$  of the optical field relative to the detection axis. With decreasing recoil along the detection axis, the two peaks of the velocity distribution draw closer together and ultimately form a single peak for  $\Theta = 90^\circ$ . Figures 9b, 9c, and 9d are fitted using Eq. (1) with the value of  $\beta$  found from fitting Fig. 9a yielding a satisfactory fit with  $R^2$  values in excess of 0.99, which provides high confidence for the asymmetry parameter we have report.



To determine the vibrational energy distribution of the ion, we have employed a two-color secondary ionization technique using time-delayed pulses. The details of this work can be found in our publication.

## Summary

We have proposed a very robust coherent optical technique, the “Stark induced adiabatic Raman passage” or SARP. Using SARP we have demonstrated population inversion between a ground vibrational ( $v=0, J$ ) and an excited ( $v > 0, J'$ ) vibrational level of  $H_2$  within the ground  $X^1\Sigma_g^+$  electronic state. In addition, we have prepared a coherent superposition of degenerate M-states belonging to a single rovibrational ( $v=1, J=2$ ) energy eigenstate within the ground  $X^1\Sigma_g^+$  electronic state of  $H_2$ . The phase entangled superposition states opened new avenues to coherently control the collision dynamics, which may also allow us to measure the geometrical phase effect in a collision process. Using SARP we have demonstrated preparation of many vibrationally excited ( $v = 1$ ) quantum states of  $H_2$  including ( $v=1, J=1, M=0, \pm 1$ ),  $H_2$  ( $v=1, J=3, M=0, \pm 1$ ),  $H_2$  ( $v=1, J=2, M=0, +2$  or  $-2$ ). A substantial population of the initial ground state was transferred to each of the prepared quantum states. For each of these state preparation we have demonstrated remarkable stability against frequency and energy fluctuations of the commercial laser pulses proving SARP to be a robust technique. To carry out sophisticated state preparation using pump and Stokes laser pulses with various polarizations, and study the collision dynamics, we have fabricated a high-vacuum reaction chamber combined with a time-of-flight spectrometer. Using the time-of-flight spectrometer we have measured the angular distributions of the photo-ions generated in a REMPI process. By using a delayed secondary ionization technique, we have also measured the internal vibrational distribution of the generated ions.

## Future goal

SARP is a powerful technique to prepare quantum states of polar and nonpolar molecules. We are currently continuing our effort to prepare higher vibrational quantum states of  $H_2$ , HD,  $D_2$ . Recently we have successfully prepared the HD ( $v=4, J=0$ ) state with nearly the complete population of the (HD ( $v=0, J=0$ )) state within the ground

electronic state. This result will be published soon. We are also developing the theory of two coherently coupled SARP processes ("Double SARP") to prepare a large ensemble into a single highly vibrationally excited quantum state close to the dissociation limit. SARP preparation of high vibrational states is especially important in studying collisions at ultralow temperature.

**Publications:**

1. N. Mukherjee and R. N. Zare, "Stark-Induced Adiabatic Raman Passage for Preparing Polarized Molecules," J. Chem. Phys. 135, 024201-1-10 (2011).
2. N. Mukherjee, W. Dong, J. A. Harrison and R. N. Zare, "Transfer of More than Half the Population to a Selected Rovibrational State of H<sub>2</sub> by Stark-Induced Adiabatic Raman Passage," J. Chem. Phys. 138,051101-1-4 (2013).
3. W. Dong, N. Mukherjee, and R. N. Zare, "Optical Preparation of H<sub>2</sub> Rovibrational Levels with Almost Complete Population Transfer," J. Chem. Phys. 139, 074204-1-6 (2013).
4. N. Mukherjee, W. Dong, and R. N. Zare, "Coherent Superposition of M-States in a Single Rovibrational Level of H<sub>2</sub> by Stark-Induced Adiabatic Raman Passage," J. Chem. Phys. **140**, 074201 (2014).
5. W. E. Perreault, N. Mukherjee and R. N. Zare, "Angular and internal state distributions of H<sub>2</sub><sup>+</sup> generated by (2+1) resonance enhanced multiphoton ionization of H<sub>2</sub> using time-of-flight mass spectrometry", J. Chem. Phys., **144**, 214201 (2016).